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H2 Vacuum UV Laser Rate Coefficients

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## ABSTRACT

Rate coefficients for electron impact excitations of  $H_2$  states relevant to vacuum UV lasers are calculated using calculated or measured dissociation cross sections. In addition, ionization and ground state vibrational excitation rate coefficients are calculated, as well as the rates of energy loss in inelastic electron-molecule collision processes in  $H_2$ . These rate coefficients are essential to calculations of laser power densities and design considerations. These rates are also needed for calculations of  $H_2$  laser produced plasmas essential to controlled fusion problems.

## PROBLEM STATUS

This is an interim report on a continuing problem.

## AUTHORIZATION

NRL Problem H02-27

Manuscript submitted March 23, 1971.

## H<sub>2</sub> VACUUM UV LASER RATE COEFFICIENTS

### INTRODUCTION

The possibility of inversion within the Lyman and the Werner bands of molecular hydrogen (H<sub>2</sub>) was first suggested by Bazhulin et al. (1). Ali and Kolb (2) followed with a feasibility study of lasing action in the Werner band and predicted ~20-kW power densities for the strongest lines. Ali (3) later generalized the calculations to include the laser emission from the Lyman band.

To calculate the laser power density and the time history of the laser pulse for each spectral line, the excitation rate coefficients to the upper laser levels must be known. Furthermore, in order to estimate the electron energy, the rate coefficients for all important inelastic electron-molecule collision processes must also be known. The purpose of this report is to calculate these rates to facilitate laser power density calculations.

In a previous report by Ali and Anderson (4), theoretical estimates (5) for the electron impact dissociation cross section of H<sub>2</sub> were utilized; however, in this report the experimentally measured dissociation cross section of Corrigan (5) will be utilized. In addition, the correct Franck-Condon factors for the Lyman band will be incorporated.

Finally, simple analytic expressions, correct to within 10-20% for most of the rate coefficients calculated in this report, will be given.

### ELECTRON EXCITATION AND IONIZATION RATE COEFFICIENTS

To calculate electron excitation or ionization rate coefficients, one must have either analytic expressions for the cross sections, or experimentally measured cross sections. If an analytic (i.e., a theoretical) expression exists, then one proceeds to average the product of this expression and the electron velocity over a Maxwellian velocity distribution. In case no such theoretical expression is available, one must rely on experimentally measured cross sections. The procedure outlined here is similar to the previous work of Ali and Anderson (6), with some refinement. As in Ref. (6), the procedure is to obtain an empirical expression for the measured cross section over the entire range of the incident electron energy. This can be done by dividing the measured cross section into a few sections, and then obtaining an empirical expression for each section as a function of the incident electron energy. Thus, if  $\sigma_{ij}$  denotes the cross section as a function of the incident electron energy  $E$ , for either the excitation or ionization of atoms, ions, or molecules, the relevant rate coefficient can be expressed as

$$X_{ij} = \langle \sigma_{ij} v \rangle = K_0 T_e^{-3/2} \int E \sigma_{ij}(E) e^{-E/T_e} dE \quad (1)$$

where  $K_0 = 6.6971 \times 10^7$ ,  $T_e$  is the electron temperature\* in units of electron volts (eV), and  $\langle \sigma v \rangle$  implies the average of  $\sigma$  with  $v$ , the electron velocity, over an electron Maxwellian velocity distribution. (For details concerning Eq. 1, see Ref. 6.)

\*Hereafter,  $T_e$  will be written as  $T$ .

Generally, one can express each of the parts of  $\sigma_{ij}$  with reasonable accuracy by one or more terms of the polynomial  $C_0 + C_1E + C_2E^2 + C_3E^3$ , or by an exponential term such as  $C_0e^{-\alpha E}$ , or by a combination of the two. Thus, the rate coefficient can now be expressed in a more general fashion as

$$X_{ij} = K_0 T^{-3/2} \sum_a \int_{E_a}^{E_{a+1}} E (C_{0a} + C_{1a}E + C_{2a}E^2 + C_{3a}E^3) e^{-[\alpha_a E + (E/T)]} dE \quad (2)$$

where  $a$  indicates the  $a$ th division of the cross section in the energy range between  $E_a$  and  $E_{a+1}$ , and the coefficients of the polynomial,  $C_{0a}$ ,  $C_{1a}$ , etc., and  $\alpha_a$  are for the  $a$ th division. The summation, of course, is over all sections of  $\sigma$ .

Typical of the integrals which arise is

$$\begin{aligned} & K_0 T^{-3/2} \int_{E_a}^{E_{a+1}} E (C_{0a} + C_{1a}E + C_{2a}E^2 + C_{3a}E^3) e^{-[\alpha_a E + (E/T)]} dE \\ &= \frac{K_0 T^{-1/2}}{1 + \alpha_a T} \left\{ e^{-E/\beta} [C_{0a}(E+\beta) + C_{1a}(E^2 + 2E\beta + 2\beta^2) \right. \\ &\quad \left. + C_{2a}(E^3 + 3E^2\beta + 6E\beta^2 + 6\beta^3) + C_{3a}(E^4 + 4E^3\beta + 12E^2\beta^2 + 24E\beta^3 + 24\beta^4)] \right\} \Big|_{E_a}^{E_{a+1}} \quad (3) \end{aligned}$$

where  $\beta = T/(1 + \alpha_a T)$ , and limits have been interchanged to eliminate the minus sign. Equation 3 is equivalent to Eq. 5 in Ref. 6. However, this new expression is in a more compact form. For example, if one has used a polynomial with only  $C_0$  and  $C_1$  as its coefficients, then in Eq. 3 one has to consider only the first two terms, i.e., the  $C_{0a}$  and  $C_{1a}$  terms.

## APPLICATION TO THE $H_2$ MOLECULE

The potential energy diagram of the  $H_2$  molecule with the relevant energy levels is shown in Fig. 1. Data from Jeppesen (7) and Herzberg (8) have been used to construct the energy diagram where the most probable (classical) excitations and radiative decays are indicated. For our purposes (laser power-density calculations in the Werner band), we need electron impact excitation rates to the C-state vibrational levels ( $v'$ ) 0, 1, 2, 3, and 4. However, the cross section given by Khare (9) is the total cross section for the excitation of the C-state without any consideration of its vibrational structure. Therefore, after obtaining the total rate for the excitation of the C-state, Spindler's (10) calculated Franck-Condon factors  $q_{v',v''}$  will be used to obtain the excitation rate for the individual vibrational level.

The total cross section  $\sigma_{XC}$  calculated by Khare is shown in Fig. 2. Following the procedure described in the preceding paragraph one can obtain the corresponding rate coefficient  $X_{XC}$  which is tabulated in Table 1. The rate coefficients for the most probable C-state vibrational levels, i.e.  $v' = 0, 1, 2, 3$ , and 4 can now be obtained by multiplying  $X_{XC}$  by the appropriate Franck-Condon factors. These rates are also shown in Fig. 3 and are given in Table 1. The Franck-Condon factors for the Werner band, as calculated by Spindler (10), are given in Table 2.

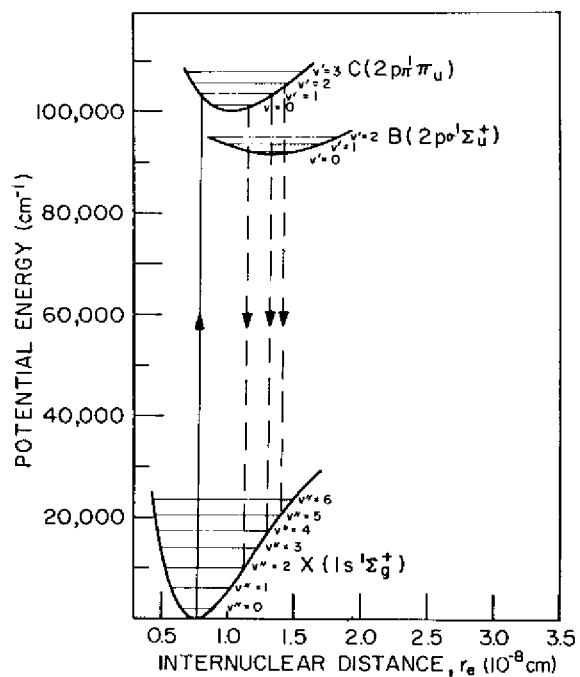


Fig. 1 - Potential energy diagram of the H<sub>2</sub> molecule indicating electronic states relevant to an H<sub>2</sub> laser.

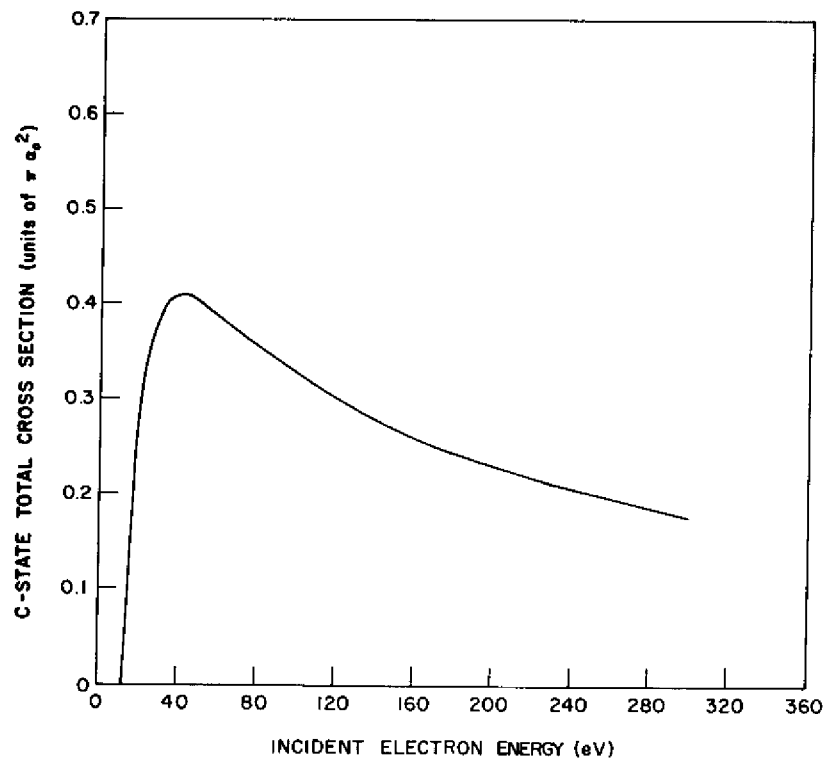


Fig. 2 - Electron impact excitation cross section  $\sigma_{XC}$  calculated by Khare (Ref. 9).



Table 1  
Excitation Rate Coefficients of the Most Probable C-State Vibrational Levels and the Total Excitation Rate Coefficient  $x_{xc}$  as a Function of the Electron Temperature T.

T	$x_{xc}$	$x_{00}^c$	$x_{01}^c$	$x_{02}^c$	$x_{03}^c$	$x_{04}^c$
1	1.86(-14)*	2.32(-15)	3.66(-15)	3.60(-15)	2.86(-15)	2.03(-15)
2	1.27(-11)	1.58(-12)	2.50(-12)	2.46(-12)	1.95(-12)	1.39(-12)
3	1.21(-10)	1.51(-11)	9.38(-11)	2.34(-11)	1.86(-11)	1.32(-11)
4	3.86(-10)	4.82(-11)	7.60(-11)	7.47(-11)	5.94(-11)	4.22(-11)
5	7.87(-10)	9.82(-11)	1.55(-10)	1.52(-10)	1.21(-10)	8.60(-11)
6	1.28(-9)	1.60(-10)	2.52(-10)	2.48(-10)	1.97(-10)	1.40(-10)
7	1.82(-9)	2.27(-10)	3.58(-10)	3.52(-10)	2.80(-10)	1.99(-10)
8	2.38(-9)	2.97(-10)	4.68(-10)	4.61(-10)	3.66(-10)	2.60(-10)
9	2.95(-9)	3.68(-10)	5.81(-10)	5.71(-10)	4.54(-10)	3.22(-10)
10	3.50(-9)	4.37(-10)	6.89(-10)	6.78(-10)	5.39(-10)	3.83(-10)
11	4.03(-9)	5.03(-10)	7.93(-10)	7.80(-10)	6.20(-10)	4.40(-10)
12	4.55(-9)	5.68(-10)	8.95(-10)	8.81(-10)	7.00(-10)	4.97(-10)
13	5.04(-9)	6.29(-10)	9.92(-10)	9.76(-10)	7.76(-10)	5.51(-10)
14	5.51(-9)	6.88(-10)	1.08(-9)	1.07(-9)	8.48(-10)	6.02(-10)
16	6.37(-9)	7.95(-10)	1.25(-9)	1.23(-9)	9.80(-10)	6.96(-10)
18	7.14(-9)	8.91(-10)	1.41(-9)	1.38(-9)	1.10(-9)	7.80(-10)
20	7.84(-9)	9.78(-10)	1.54(-9)	1.52(-9)	1.21(-9)	8.57(-10)
22	8.46(-9)	1.06(-9)	1.66(-9)	1.64(-9)	1.30(-9)	9.25(-10)
24	9.02(-9)	1.13(-9)	1.78(-9)	1.75(-9)	1.39(-9)	9.86(-10)
26	9.52(-9)	1.19(-9)	1.87(-9)	1.84(-9)	1.47(-9)	1.04(-9)
28	9.98(-9)	1.25(-9)	1.96(-9)	1.93(-9)	1.54(-9)	1.09(-9)
30	1.04(-8)	1.30(-9)	2.05(-9)	2.01(-9)	1.60(-9)	1.14(-9)

\*Numbers in parentheses indicate the power of 10 by which the entries are Multiplied.

#### EXCITATION RATE COEFFICIENT $x_{xb}$

In a similar way, the expression  $x_{xb}$  is obtained for the excitation rate coefficient to the B electronic state using the calculated (9) cross section shown in Fig. 4.

The excitation of the B-state vibrational levels becomes of considerable interest for the calculations of the Lyman band laser lines in the light of the experimental observation and proof (11) of laser action within this band. The calculations of Lyman band laser emission intensities by Ali (12) will be published in the near future. The appropriate Franck-Condon factors for the Lyman band are given in Table 3.

The excitation rate coefficient  $x_{xb}$  is given in Table 4 along with the excitation rate coefficients of some of its vibrational levels. These results are also shown in Fig. 5.

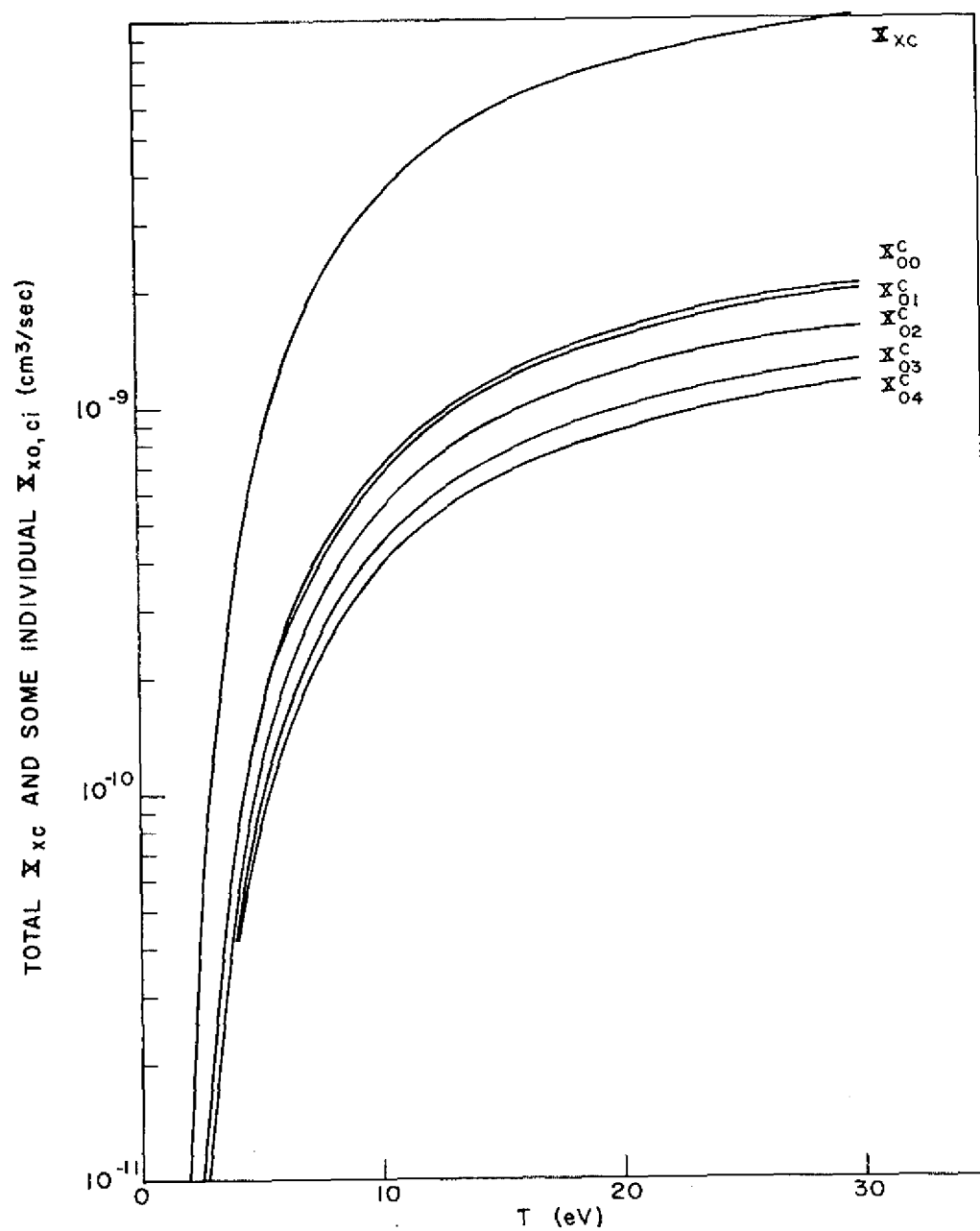


Fig. 3 - Electron impact excitation rate coefficient  $X_{xc}$  and the rate coefficients of the most probable C-state vibrational levels.

Table 2  
The Franck-Condon Factors for the Most Probable Excitations and Possible  
Laser Transitions\* in the Werner Band

v'	v''								
	0	1	2	3	4	5	6	7	8
0	0.1248	0.3254	0.3318	0.1679	—	—	—	—	—
1	0.1968	0.1397	—	0.2138	0.2900	0.1306	—	—	—
2	0.1936	—	0.1157	—	—	0.2824	0.2255	—	—
3	0.1539	—	0.1044	—	0.1319	—	0.1934	0.3	—
4	0.1093	—	—	—	—	—	—	—	0.3479

\*Transitions with Franck-Condon factors below 0.1 are neglected.

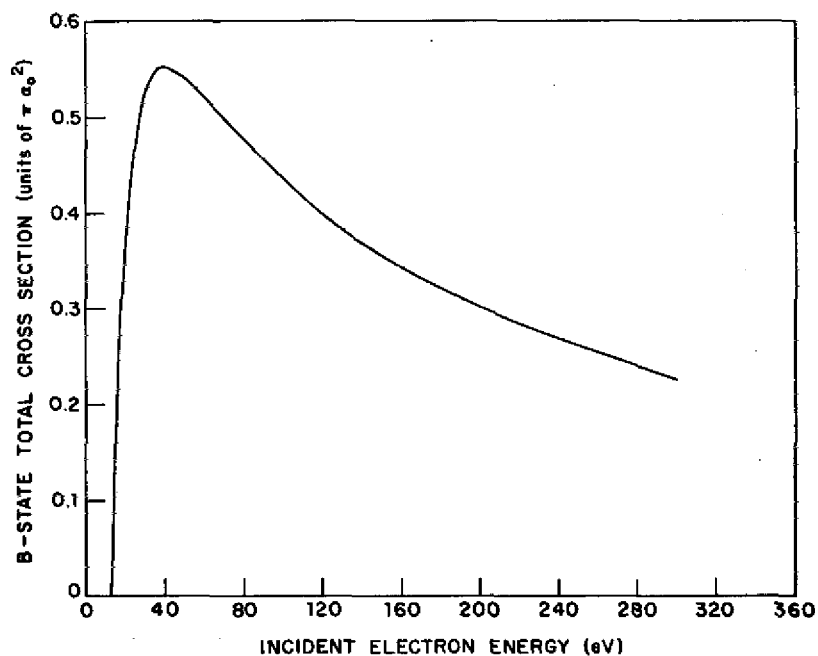


Fig. 4 - Electron impact excitation cross section  $\sigma_{XB}$  calculated by Khare.

Table 3  
Franck-Condon Factors for  $H_2$  Band Systems

		$(B^1\Sigma_u^+ - X^1\Sigma_g^+)$					
$v'$ \ $v''$	0....	9	10	11	12	13	14
2	0.0273....	0.2552	0.1251				
3	0.0427....		0.2952	0.1693			
4	0.0563....			0.3442	0.1600		
5	0.0663....				0.4347		
6	0.0723....					0.4366	
7	0.0740....					0.2600	
8	0.0725....						0.2781
9	0.0688....						
10	0.0634....						
11	0.0575....						0.1546
12	0.0513....						
13	0.0452....						
14	0.0394....						

It was stated earlier that one only has to multiply  $x_{XB}$  by the appropriate Franck-Condon factors to obtain the excitation rate coefficient to any desired B-state vibrational level.

#### IONIZATION RATE COEFFICIENT $x_i$

The ionization rate coefficient  $x_i = \langle \sigma_i v \rangle$  was obtained in the same way as discussed earlier using a measured (13) cross section shown in Fig. 6.

$x_i$  is shown in Fig. 7 and given in Table 5.

#### GROUND STATE VIBRATIONAL EXCITATION RATE COEFFICIENT $\langle \sigma_v v \rangle$

The measured electron impact excitation cross section of the ground state vibrational levels of  $H_2$  has been measured by Schulz (14). This cross section which is shown in Fig. 8 is mainly the cross section of the first vibrational level of  $H_2$ . The rate coefficient  $\langle \sigma_v v \rangle$  is shown in Fig. 9 and tabulated in Table 6.

#### EXCITATION RATE COEFFICIENT OF THE TRIPLET STATES OF $H_2$ (THE DISSOCIATION RATE)

The hydrogen molecule has many triplet states. There are at least nineteen known such states, intercombination transitions being forbidden. Therefore, the electron impact excitation of any triplet state will eventually lead to the dissociation of  $H_2$ . The known triplet states are bound electronic states, except for one which is a repulsive state and leads to direct dissociation of the molecule. This state is  $^3\Sigma_u^+$ , and consequently dissociation results. The cross section for electron impact dissociation of  $H_2$ , shown in Fig. 10,

Table 4  
Excitation Rate Coefficients of the Most Probable  
B-State Vibrational Levels and the Total Excitation  
Rate Coefficient  $X_{XB}$  as a Function of the Electron  
Temperature T.

T	$X_{XB}$	$X_{04}^B$	$X_{05}^B$	$X_{06}^B$
1	2.09(-14)*	1.18(-15)	1.38(-15)	1.5 (-15)
2	1.50(-11)	8.45(-13)	9.94(-13)	1.08(-12)
3	1.48(-10)	8.33(-12)	9.81(-12)	1.07(-11)
4	4.85(-10)	2.73(-12)	3.21(-11)	3.50(-11)
5	1.01(-9)	5.68(-11)	6.69(-11)	7.30(-11)
6	1.65(-9)	9.28(-11)	1.09(-10)	1.19(-10)
7	2.37(-9)	1.33(-10)	1.57(-10)	1.71(-10)
8	3.12(-9)	1.75(-10)	2.06(-10)	2.25(-10)
9	3.88(-9)	2.18(-10)	2.57(-10)	2.80(-10)
10	4.63(-9)	2.60(-10)	3.06(-10)	3.35(-10)
11	5.35(-9)	3.01(-10)	3.54(-10)	3.87(-10)
12	6.05(-9)	3.41(-10)	4.01(-10)	4.37(-10)
13	6.71(-9)	3.78(-10)	4.44(-10)	4.85(-10)
14	7.35(-9)	4.14(-10)	4.87(-10)	5.31(-10)
16	8.51(-9)	4.79(-10)	5.64(-10)	6.15(-10)
18	9.56(-9)	5.38(-10)	6.34(-10)	6.91(-10)
20	1.05(-8)	5.91(-10)	6.96(-10)	7.59(-10)
22	1.13(-8)	6.36(-10)	7.49(-10)	8.16(-10)
24	1.21(-8)	6.81(-10)	8.02(-10)	8.75(-10)
26	1.28(-8)	7.2 (-10)	8.48(-10)	9.25(-10)
28	1.34(-8)	7.54(-10)	8.88(-10)	9.68(-10)
30	1.39(-8)	7.82(-10)	9.21(-10)	1.00(-10)

\*Numbers in parentheses indicate the power of 10 by which the entries are multiplied.

which is the sum of all triplet state excitations, has been measured by Corrigan(5). The corresponding rate coefficient is given in Table 7 and shown in Fig. 11.

#### RATE OF ENERGY LOSS BY ELECTRONS TO THE MOST IMPORTANT INELASTIC COLLISIONS IN $H_2$

The electrons lose their energies via inelastic collisions. Among the most important processes are (a) ionization, (b) excitation of electronic states, (c) dissociation, and (d) excitation of the ground state vibrational levels. The rate coefficients for these processes were calculated in the preceding sections. To obtain the rate of energy loss by electrons to the above processes, one must multiply the rate by the corresponding threshold

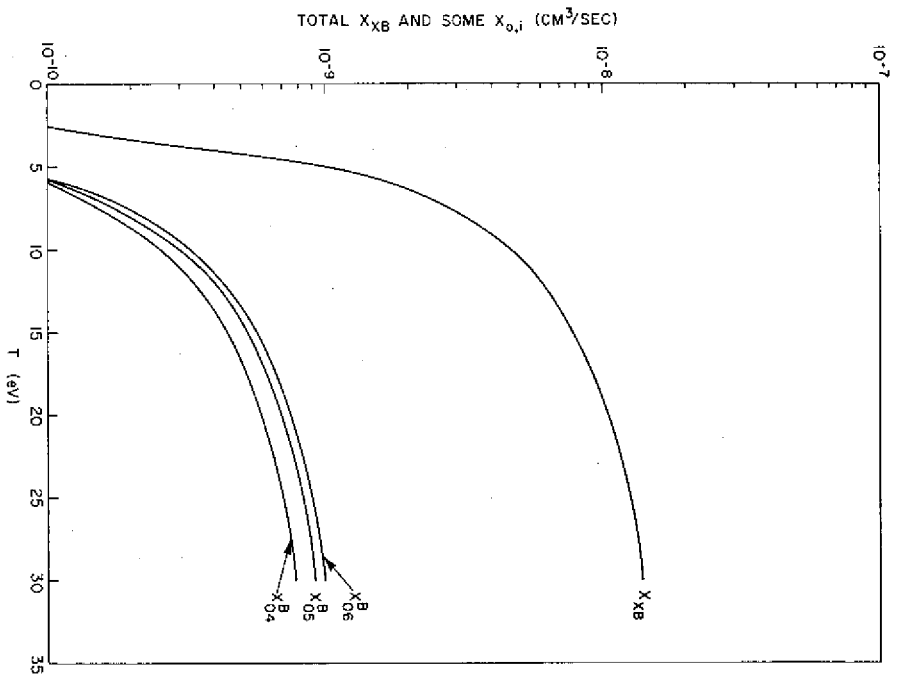


Fig. 5 - Electron impact excitation rate coefficient  $X_{XB}$  and the rate coefficients of the most probable B-state vibrational levels.

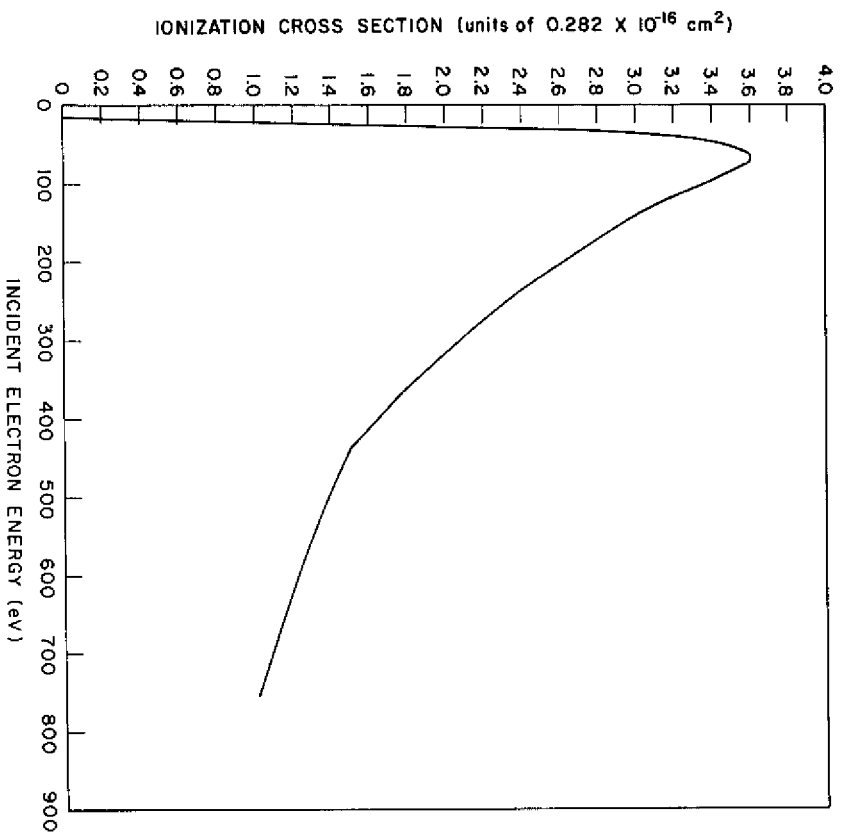


Fig. 6 - Molecular  $H_2$  ionization cross section measured by Tate and Smith (13) as a function of the incident electron energy.

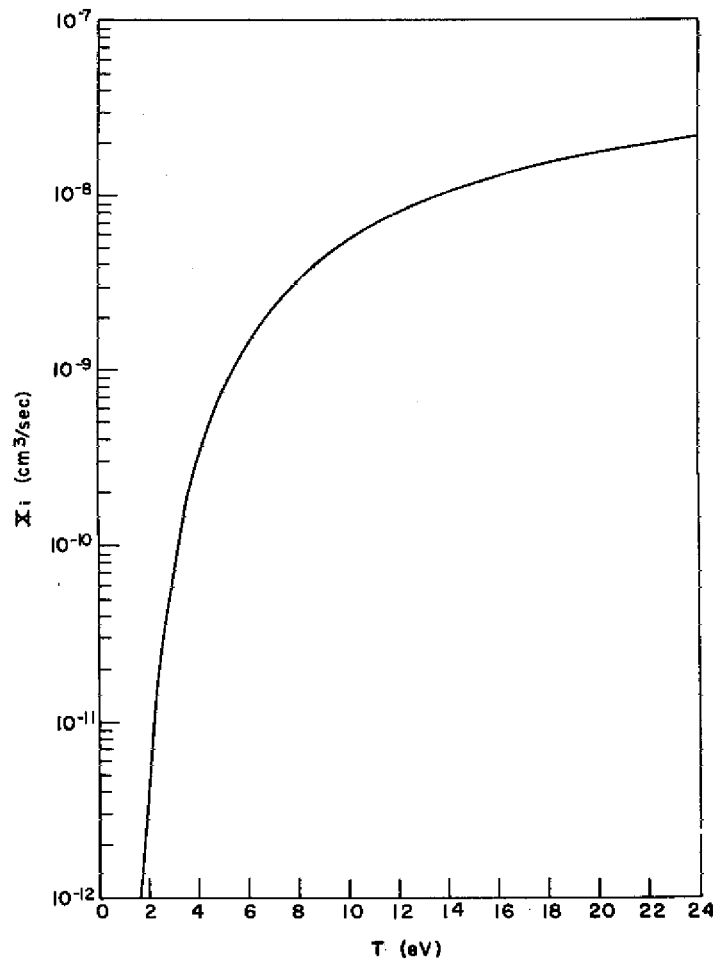


Fig. 7 - Ionization rate coefficient  $X_i$ .

energy. However, for B- and C-state excitation energies, one must take an average energy in each case. The average energy can be obtained by multiplying the excitation energy of each vibrational level by the corresponding Franck-Condon factor. Accordingly one has for the average energies of the B and C states, 12.28 eV and 12.82 eV, respectively. Figure 12 shows the rates of electron energy loss to the most important processes.

#### SIMPLE EXPRESSIONS FOR THE RATE COEFFICIENTS

Least-squares techniques have been used to fit an expression of the form

$$T^{-1/2} \exp \left( -E_{th}/T \right) \left( B_0 + b_1 T + b_2 T^2 \right) \quad (4)$$

to the various rate coefficients, where  $E_{th}$  is the appropriate threshold energy. In general this expression fits to within a few percent over most of the energy range of interest. Table 7 lists  $E_{th}$ ,  $b_0$ ,  $b_1$ ,  $b_2$ , and the temperature range  $T_1 \leq T \leq T_2$  over which Eq. 4 fits the rate coefficient to within 20%.

Table 5  
Ionization Rate Coefficient  
 $x_i$  of the  $H_2$  Molecule as a  
Function of the Electron  
Temperature  $T$ .

$T$	$x_i$
1	$3.16(-15)^*$
2	$5.56(-12)$
3	$7.82(-11)$
4	$3.18(-10)$
5	$7.76(-10)$
6	$1.45(-9)$
7	$2.31(-9)$
8	$3.31(-9)$
9	$4.42(-9)$
10	$5.60(-9)$
11	$6.83(-9)$
12	$8.09(-9)$
13	$9.35(-9)$
14	$1.06(-8)$
16	$1.31(-8)$
18	$1.55(-8)$
20	$1.77(-8)$
22	$1.98(-8)$
24	$2.18(-8)$
26	$2.36(-8)$
28	$2.53(-8)$
30	$2.70(-8)$

\*Numbers in parentheses  
indicate the power of 10 by  
which the entries are to be  
multiplied



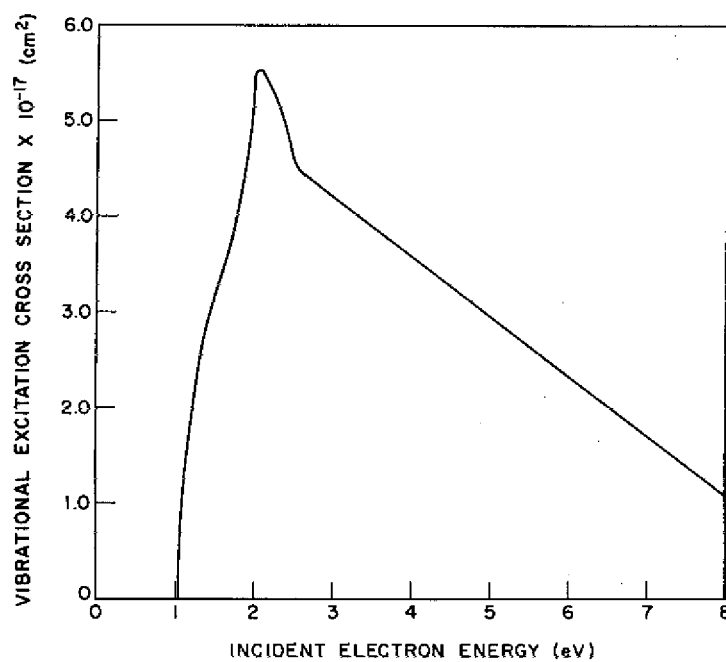


Fig. 8 - Electron impact excitation cross section of the ground-state vibrational levels measured by Schulz (14) as a function of the incident electron energy.

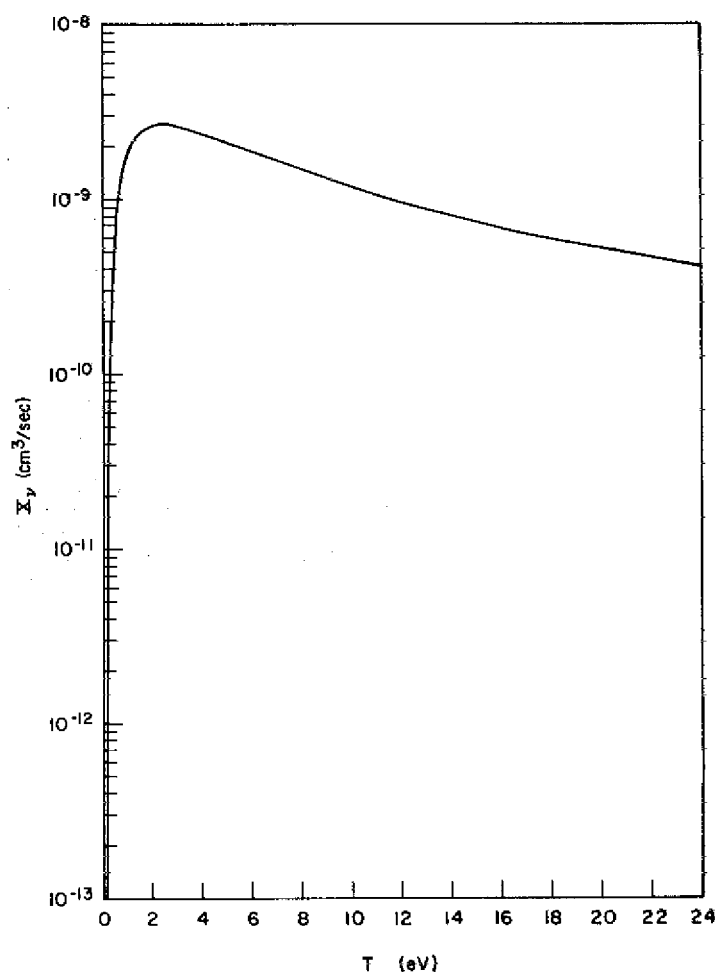


Fig. 9 - Computer calculation of the rate coefficient  $\langle \sigma_v v \rangle$ .

Table 6  
Vibrational Excitation Rate  
Coefficient  $\langle \sigma_v v \rangle$  as a  
Function of the Electron  
Temperature T.

T	$\langle \sigma_v v \rangle$
1	1.75(-9)
2	2.66(-9)
3	2.65(-9)
4	2.40(-9)
5	2.12(-9)
6	1.87(-9)
7	1.65(-9)
8	1.46(-9)
9	1.31(-9)
10	1.18(-9)
11	1.06(-9)
12	0.966(-9)
13	0.884(-9)
14	0.812(-9)
16	0.694(-9)
18	0.601(-9)
20	0.527(-9)
22	0.466(-9)
24	0.418(-9)
26	0.375(-9)
28	0.341(-9)
30	0.312(-9)

Fig. 10 - Electron impact dissociation  
cross reaction of  $H_2$ .

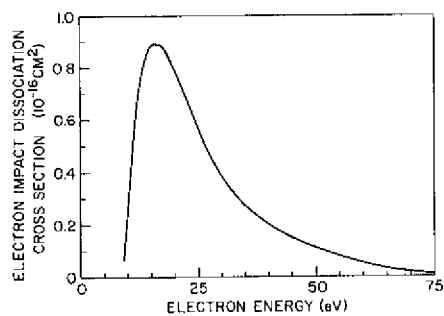


Table 7  
Least-Squares Parameters for Excitation Rates

Excitation State	Range(eV)	$E_{th}$	$b_0$	$b_1$	$b_2$
$b^3\Sigma_u$	$3.2 \leq T \leq 24.0$	8.8	$1.653(-8)^*$	$8.955(-10)^*$	$-3.434(-11)^*$
$e + H_2 \rightarrow H + H + e$	$3.3 \leq T \leq 24.0$	8.8	$4.365(-8)$	$3.369(-9)$	$-1.133(-10)$
$X_i$	$3.9 \leq T \leq 24.0$	15.43	$-1.479(-8)$	$1.032(-8)$	$-5.158(-11)$
$X_{0C}$	$1.3 \leq T \leq 24.0$	12.82	$5.006(-9)$	$3.866(-9)$	$-3.925(-11)$
$X_{0B}$	$1.7 \leq T \leq 24.0$	12.28	$1.070(-9)$	$5.453(-9)$	$-5.817(-11)$

\*Numbers in parentheses indicate the power of 10 by which the entries are to be multiplied.

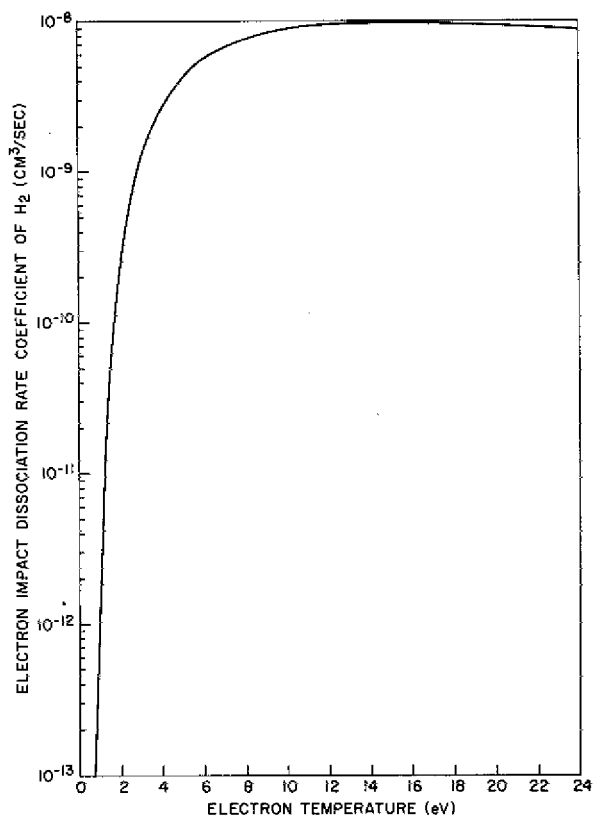


Fig. 11 - Electron impact dissociation rate coefficient of  $H_2$ .

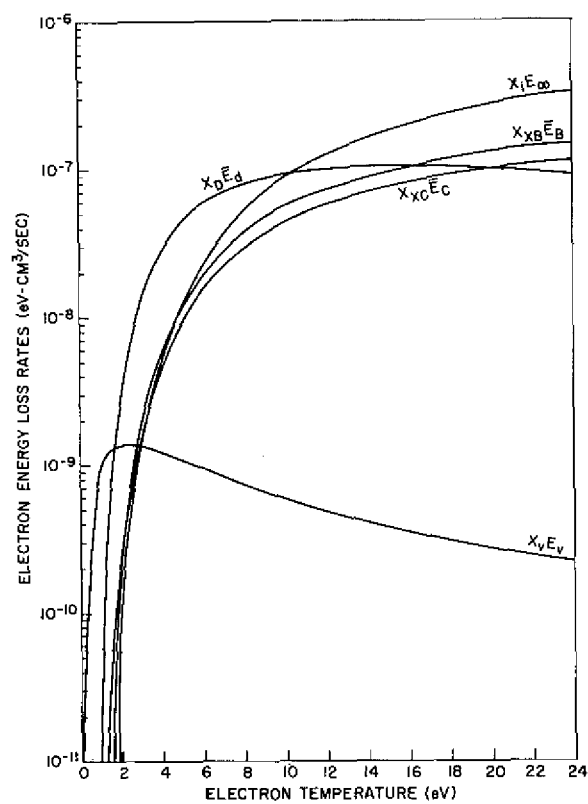


Fig. 12 - Electron energy loss rates to the most important processes as a function of electron temperature.

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